

DEVELOPMENT OF THE METHOD OF HIGH DENSITY FUEL COMMINUTION BY HYDRIDE-DEHYDRIDE PROCESSING.

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ABSTRACT

The report contains the brief review of the existing methods for high density uranium alloys powder production. The results of researchers on comminution of U-Mo alloys by hydride-dehydride processing are presented. As a result of work it is shown that this method is suitable for comminution of U-Mo alloys. It provides the production of fuel grains of needed size, structure and phase composition with high efficiency. The investigations of interaction between U-Mo fuel grains and matrix material are being carried out now. The preliminary results has shown that compatibility of U-Mo powder manufactured by hydride-dehydride processing with matrix material is similar (not worse) to that of the fuel grains produced by other methods.

INTRODUCTION

At present time the set of new high density uranium alloys is under investigation in the frames of RERTR program [1], among which there are U-Mo alloys with different Mo content. To manufacture dispersion fuel elements (FEs) on the basis of these alloys it is necessary to develop effective powder production methods.

Among the number of existing powder production methods there are several most perspective ones which can be suitable for high density uranium alloys: mechanical crushing (milling, grinding, etc); mechanical crushing with preliminary alloying to increase brittleness; cryogenic mechanical crushing; atomization, as well as hydride-dehydride(HD) processing.

Each of above-mentioned methods has some advantages and shortcomings briefly set fourth below in the item 1.1.

The article contains the results of investigations carried out in GNC RF VNIINM on studying the possibility of U-Mo powder production by hydride-dehydride processing of alloy.

1. DEVELOPMENT OF THE METHOD OF U-MO POWDER PRODUCTION BY HYDRIDE-DEHYDRIDE PROCESSING OF ALLOY.

1.1 Some issues of uranium alloys powder production.

High ductility of majority high density uranium alloys excludes, practically, the usage of mechanical crushing (comminution) because of the low efficiency and high labor-consumption. But, for example,

in combination with preliminary alloying, aimed at formation of microstructure containing brittle phases, such approach makes it possible to reduce labor-consumption meanwhile losing a bit in density.

As far as ductile uranium alloys are concerned atomization seems to be the most promising powder production option. For example the method of atomization of the melt developed in KAERI [2], or the method of arc-plasma atomization, which is used in some countries including Russia [3]. Nevertheless there are some drawbacks inherent to it, which are associated with some limitations in applicability for FE production due to spherical shape of fuel particles. The spherical shape of powder particles makes difficult to manufacture thin-walled FEs because of possible inculcation into thin cladding and danger of contact with coolant.

HD processing is free of foregoing drawbacks and at the same time has some advantages: complicated and power-consuming equipment is not necessary; the process is simple enough and allows to produce granules of irregular shape and of wide size range.

Taking into account the above-mentioned information an attempt was undertaken to produce U-Mo powder by HD processing. Despite the fact that HD processing is used in some nuclear technologies [4], there is no information until today about successful attempts on using HD processing for U-Mo powder production.

1.2. Development of laboratory-scale method of U-Mo powder production by hydride-dehydride processing.

It is well-known [5], that U-Mo dispersion fuel in metastable γ -phase state is considered as a perspective fuel in frames of RERTR program due to it's high irradiation stability and corrosion resistance.

The investigations of the possibility of γ -phase U-Mo powder production by HD processing are carrying out now in GNC RF VNIINM. As a result of these R&D activities, the laboratory-scale method, which makes it possible to manufacture the fuel particles of needed size and phase composition, was developed. Well-known technological procedures form the basis of this method (figure 1) [6,7]:

- heat treatment of alloy to decompose U-Mo metastable γ -phase;
- HD processing to comminute U-Mo alloy;
- Heat treatment to recover U-Mo metastable γ -phase.

Thus the lab-scale method of U-Mo powder production represents the HD processing of initial U-Mo alloy, preliminary heat treated to decompose metastable γ -phase into α U and γ^1 -phase. HD processing of two- phase alloy results in fuel particle formation. After completion of HD processing a quenching

of fuel particles from γ -phase area of U-Mo diagram has to be conducted to recover the initial γ -phase state.

2. OUT-OF-PILE INVESTIGATIONS OF FUEL PARTICLES MANUFACTURED BY HYDRIDE-DEHYDRIDE PROCESSING.

2.1 Powder size distribution research

The size of fuel particles for dispersion FEs of different type research reactors lies as usual in the range from 50 μm to 200 μm [8]. Consequently, the first issue to be solved during the development of U-Mo powder production method is to manufacture fuel particles of needed size. As a result of researches performed, the dependence of fuel particles size distribution upon the duration of heat treatment before HD processing was established. The results of sieve analyses are given in figure 2. As it can be seen (fig. 2), the increasing of the time of heat treatment leads to increasing of weight share of smaller size fuel particles. Heat treatment during 12 hours provides high weight share (78.7%) of fuel particles of needed size.

2.2 Phase composition of U-Mo powder produced by HD processing.

The second key issue associated with U-Mo powder production by HD processing is to provide the same phase composition in the powder as for as-cast state. U-Mo(6.5wt%) alloys melted in induction furnace with graphite crucible and graphite mould were found to be metastable b.c.c. γ -phase with the lattice parameter $a=3.439 \text{ \AA}$ (fig. 3.1).

Before HD processing the alloy is heat treated to decompose the metastable γ -phase. As a result of this decomposition U-Mo alloy is transformed into two-phase state: α -U and γ^1 -phase with lattice parameter $a^1=3.417 \text{ \AA}$ (fig. 3.2)

After HD processing, the U-Mo powder is in two-phase state with the same lattice parameters as before HD. Quenching of such powder from the γ -phase area leads to the transition into single-phase state: γ -phase with lattice parameter $a=3.441 \text{ \AA}$ (fig. 3.3), which is practically the same as for as-cast state. It means that quenching after HD processing results in recovering of γ -phase state in the U-Mo powder which coincides with that in U-Mo alloy. This can be seen with X-ray diffraction patterns on figure 3.

At the same time it is necessary to notice, that half-width [$\Delta(2\theta)$] of X-ray peaks is higher at the X-ray pattern of U-Mo powder (fig 3.3) in comparison with U-Mo alloy (fig.3.1).

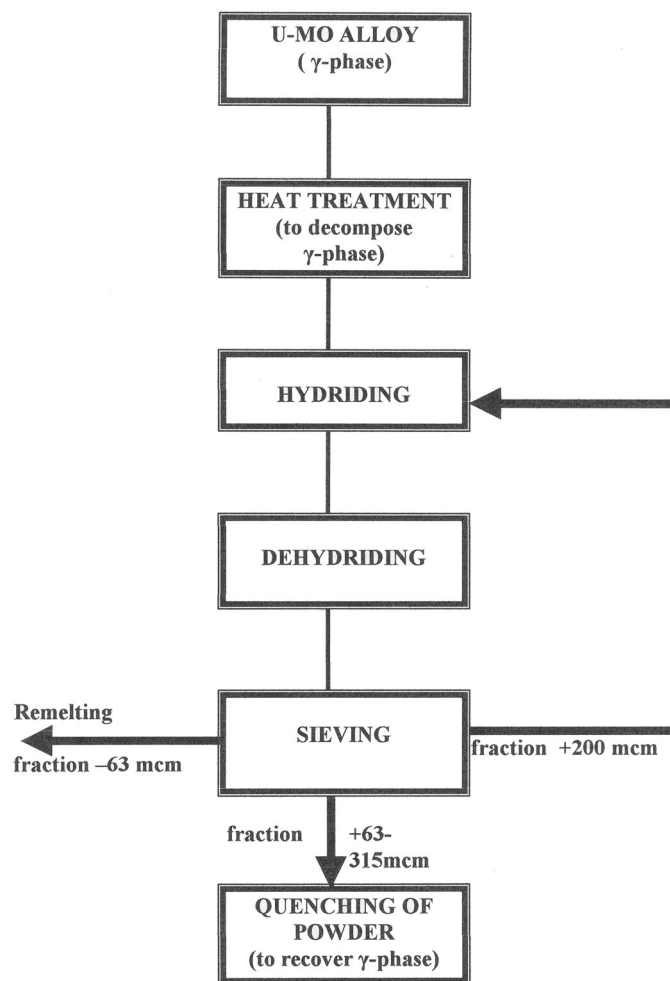
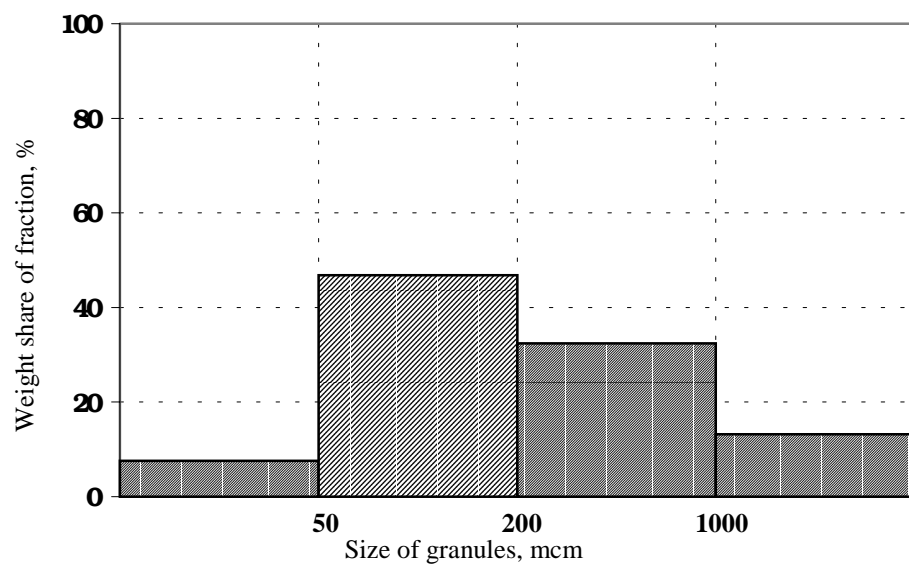
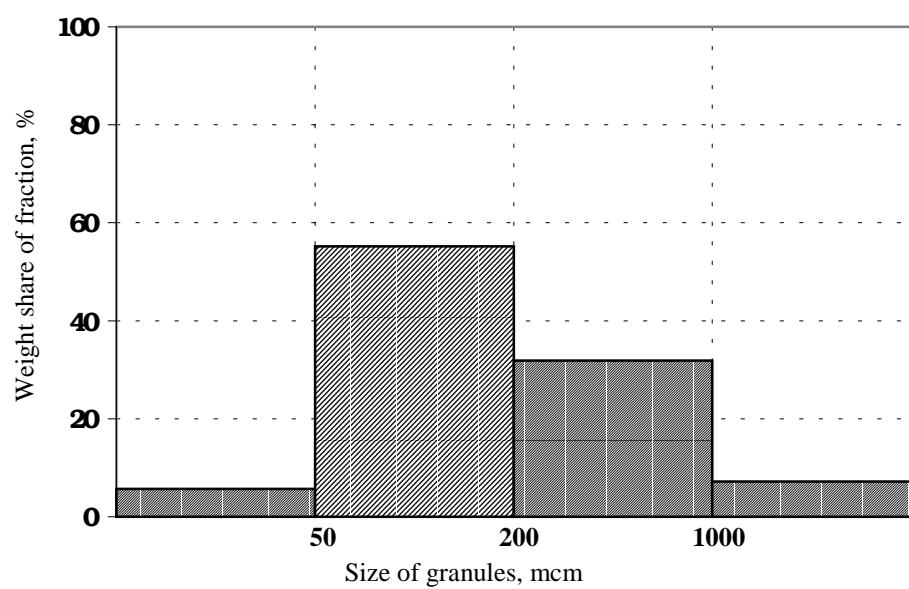


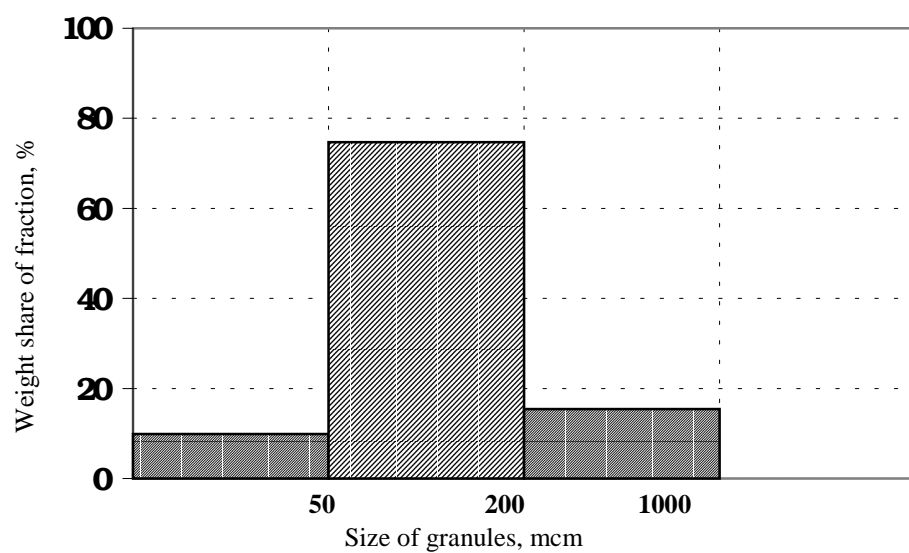
Figure.1. Technological flowsheet of U-Mo powder production method by hydride-dehydride processing.



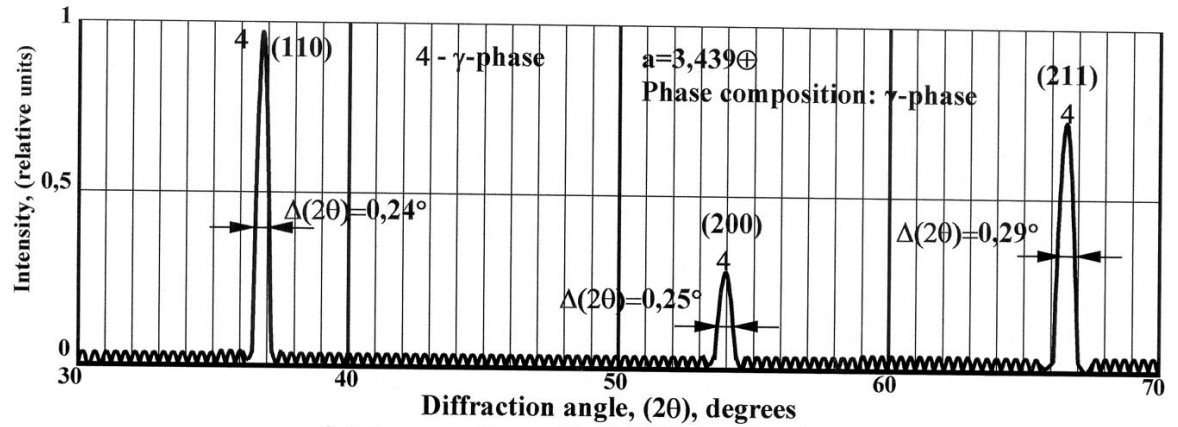
a) Duration of heat treatment 3 hours



b) Duration of heat treatment 6 hours



c) Duration of heat treatment 12 hours



3.1 X-ray pattern of initial U-Mo alloy (as-cast)

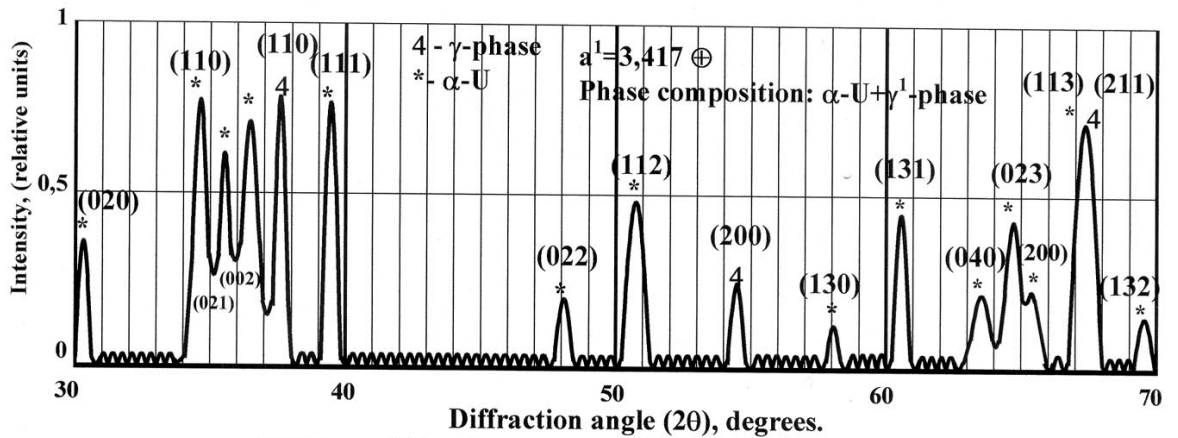
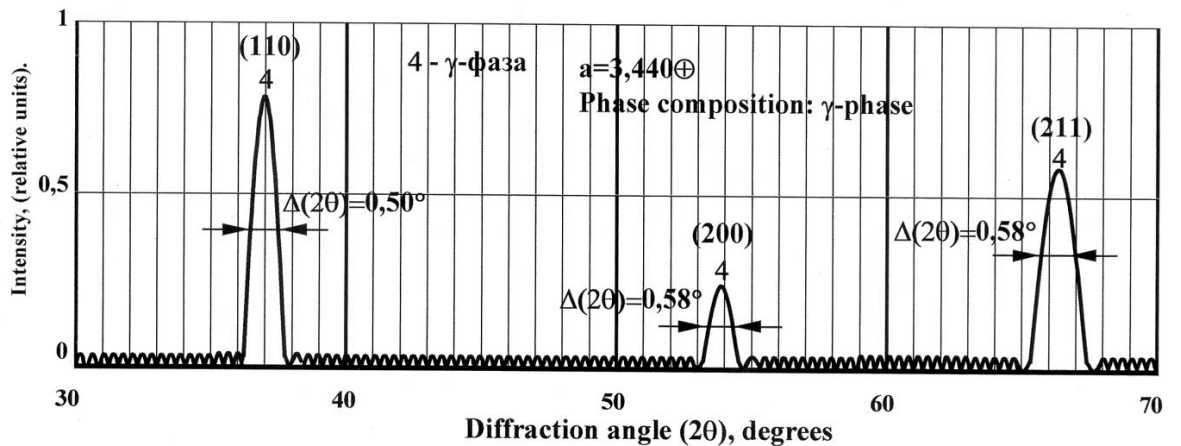
3.2 X-ray diffraction pattern of U-Mo alloy (as-heat treated to decompose γ -phase)3.3 X-ray diffraction pattern of U-Mo powder, manufactured by HD processing, (as-quenched from γ area)

Fig. 3 X-ray diffraction patterns of U-Mo (6,5wt%) alloy at different stages of powder production method with HD processing.

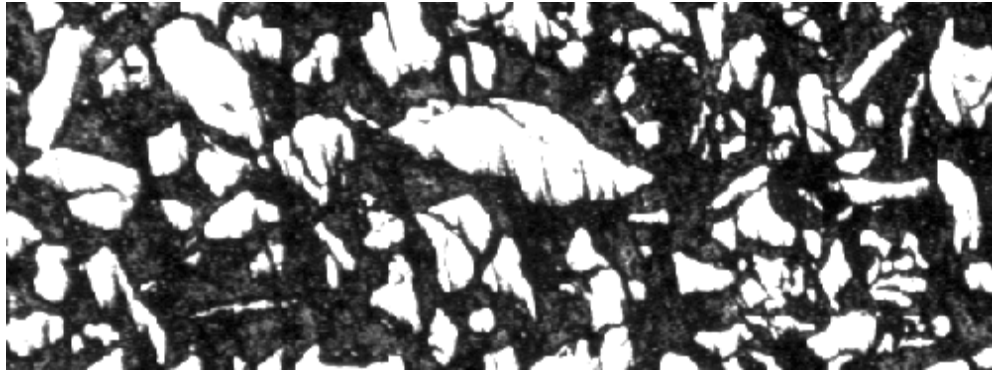
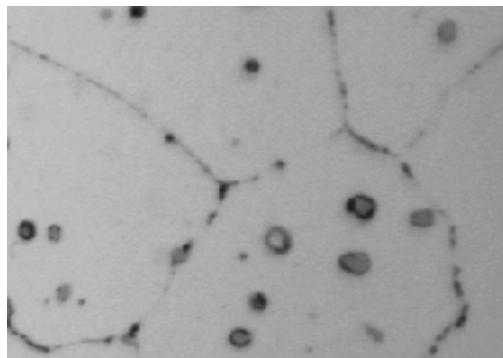
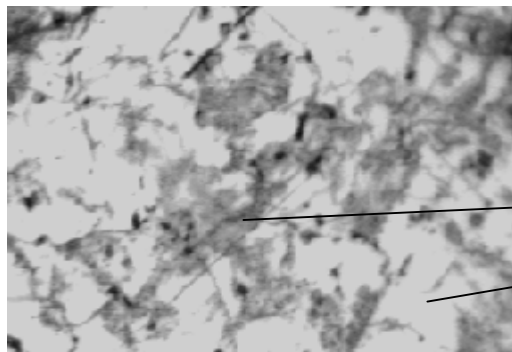


Fig.4 Cross-section of U-Mo granules produced by HD processing (x50)



a) as-cast microstructure
(x500)



b) microstructure of granules
after HD processing (x500)

α -U (0.5wt% Mo)

γ -phase

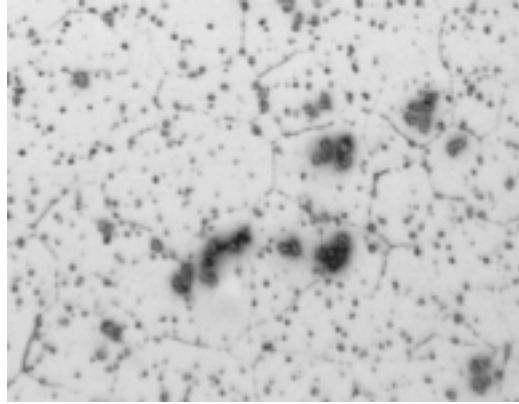


Fig.5 Microstructure of U-Mo(6.5wt%) alloy at different stages of HD processing

2.3 Microstructure of U-Mo powder produced by HD processing.

The shape of U-Mo granules produced by HD processing is “irregular”, that can be seen in the figure 4. Microstructure of U-Mo granules is given in the figure 5 as well as microstructure of as-cast specimen (fig 5a). Both microstructures are practically identical with the exception of grain size, which makes ~60 mcm for as-cast specimen and ~ 4 mcm ÷ 12 mcm for as-quenched granules, and represent single γ -phase. It means that HD processing retains, on the whole, the initial microstructure of as-alloyed specimen.

3. Discussion.

It is known, that variation of lattice parameter of γ -phase type U-Mo alloys depends upon the quantity of Mo solved in U and is described by Vegardt's law [9]:

$$a = a_0 - b_0 \cdot C,$$

where - a – lattice parameter of γ -phase alloy;

C – Mo content in alloy;

a_0 , b_0 – constants ($a_0 \cong 3,481 \text{ \AA}$, $b_0 \cong 0,003 \text{ \AA/at.}\%$).

In such case, for example, the lattice parameter of U-6.5wt%Mo alloy has to be equal to 3.441 \AA , that nearly coincides with measured one of as-cast specimen – 3.439 \AA and of U-Mo powder produced by HD processing – 3.440 . At the same time, Mo content in γ^1 -phase ($a^1 = 3.417 \text{ \AA}$) of as-heat treated U-Mo alloy will make – 9.8wt% in accordance with Vegardts equation. These data may be used to explain the observed broadening of diffraction peaks at X-ray patterns (fig.3) as a result of not only fine sub-grains and micro-stresses, but also of non-uniform distribution of Mo in uranium lattice. This situation may take place because of measured variations of Mo content in uranium lattice at different stages of powder production process.

Such peculiarities of microstructure and phase content may influence the interaction at the boundary “fuel particle – matrix Al” of dispersion composition. Consequently it is necessary to perform additional studies of compatibility of U-Mo particles with Al matrix in dispersion fuel element. Such work is being executed now and in accordance with preliminary results the compatibility is not worth than for U-Mo particles produced by another methods.

CONCLUSIONS

- The possibility of U-Mo powder production by HD processing is studied
- The preliminary parameters of HD method for U-Mo powder production are determined. It is shown that HD processing is effective option to manufacture U-Mo fuel particles of needed size, phase composition and structure.

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